

### **REMARKS**

Applicant thanks the Examiner for carefully considering this application. Please reconsider the application in view of the above amendments and the following remarks.

#### **Disposition of Claims**

Claims 1, 2, 5, 18, and 19 are pending. Claim 1 is independent. The remaining claims depend from claim 1.

#### **Rejections over U.S. Patent No. 5,178,766**

Claims 1, 2, 5, 18 and 19 stand rejected under 35 U.S.C. § 102(b) as being anticipated or, in the alternative, obvious over U.S. Patent No. 5,178,766 (Ikeda). This rejection is respectfully traversed.

The claimed invention generally relates to a composite reverse osmosis membrane having a high salt rejection and a high permeate flow rate. As recited in claim 1, a reverse osmosis membrane in accordance with the invention is prepared by a method comprising the steps of: (1) forming a layer on a porous support by coating a solution A comprising an amino compound having at least two reactive amino groups; (2) contacting the layer with a solution B comprising a polyfunctional acid halide compound; and (3) subsequently contacting the layer with a solution C comprising the polyfunctional acid halide compound at a higher concentration than a concentration of the solution B to form the polyamide skin layer. The polyamide layer thus formed is characterized in that a contact angle between a surface of the polyamide skin layer and water is no more than 45°, sodium chloride rejection is at least 98%, and a permeate flow rate is at least 0.7

$\text{m}^3/\text{m}^2\cdot\text{day}$  when evaluated by using feed water which has pH 6.5, 0.05 weight % of salt, an operation pressure of  $5 \text{ kgf}/\text{cm}^2$  and a temperature of  $25^\circ\text{C}$ .

Advantageously, a reverse osmosis membrane in accordance with the invention is more hydrophilic (as evidenced by smaller water contact angles,  $< 45^\circ$ ) and provides high salt rejection (at least 98%) and high flow rate (at least  $0.7 \text{ m}^3/\text{m}^2\cdot\text{day}$ ). The flow rate is evaluated using feed water which has pH 6.5, 0.05 weight % of salt, an operation pressure of  $5 \text{ kgf}/\text{cm}^2$  and a temperature of  $25^\circ\text{C}$ .

As noted above, claim 1 requires a water contact angle is no more than  $45^\circ$ . As shown in Table 1, on page 14 of the specification, a low water contact angle is a good indication of high salt rejection and high flux. Reverse osmosis membranes (Examples 2 and 3) prepared according to processes recited in claim 1 have significantly lower water contact angles ( $<45^\circ$ ) than that ( $51^\circ$ ) of a membrane prepared according to a prior art two-solution method (Comparative Example 1) or that ( $47^\circ$ ) of a membrane prepared according to a three-solution method, in which Solutions B and C have identical concentrations (Comparative Example 2).

By using a three-solution method, in which Solution C has a higher concentration than that of Solution B, embodiments of the invention produce membranes that have lower water contact angles (i.e., higher hydrophilicity). While not wishing to be bound by the mechanism by which this desired property is obtained, it is hypothesized that the additional Solution C at a higher concentration is able to react with the remaining amino groups that were not accessible to the polyfunctional acid halides in Solution B. As a result, the three-solution approach as described in the present invention is able to produce more amide linkages and/or carboxylic groups (hence, higher hydrophilicity) than the

prior art approach.

In contrast, the Applicant respectfully asserts that the chemistry disclosed by Ikeda to form a reverse osmosis membrane does not yield a membrane having the properties claimed above. As evidence, the Applicant submits a declaration, pursuant to 37 C.F.R. §1.132 (the "Declaration") that provides additional support the Applicant's argument that Ikeda does not teach all of the features claimed in the present invention.

As set forth in the Declaration, in Ikeda, the performance of a reverse osmosis membrane is evaluated by using a 1500 ppm NaCl solution at an operation pressure of 15 kgf/cm<sup>2</sup>. In contrast, the performance of a reverse osmosis membrane in accordance with embodiments of the present invention was tested at 500 ppm at an operation pressure of 5 kgf/cm<sup>2</sup>. As shown in the Declaration (referring to the table on page 2), by way of reference to comparative example 1 and example 1, increasing the amount of NaCl in the solution and increasing the operation pressure necessarily have the effect of increasing the salt rejection rate (simply because more salt is present, at a higher pressure--making it less likely to pass through).

Consequently, although the flux of a reverse osmosis membrane disclosed in Ikeda appears to fall within the range of the present invention, its NaCl rejection is lower than the range of the present invention when converted to a value corresponding to the test conditions of the present invention. The Declaration shows that the NaCl rejection for comparative example 1 is 97% when measured under the conditions disclosed by Ikeda (as opposed to 93% when tested under the conditions used by the present invention). As the following mathematical discussion shows, these tested results empirically correlate with the Applicant's predicted value for Ikeda's membranes if

tested under the Applicant's conditions.

In reverse osmosis, flux and salt permeation can be represented by the following equations:

$$J_v = L_p (\Delta P - \Delta \pi) \quad (1)$$

$$J_s = B (C_2 - C_3) \quad (2)$$

where  $J_v$  is the volume flow rate (flux),  $L_p$  is the coefficient of water permeation,  $\Delta P$  is the difference in pressures across the membrane,  $\Delta \pi$  is the difference in osmotic pressures across the membrane,  $J_s$  is the solute flux,  $B$  is the coefficient of solute permeation,  $C_2$  is the solute concentration at the surface of the membrane (i.e., the concentration in the feed solution), and  $C_3$  is the solute concentration in the permeate (the solution that has permeated through the membrane).

Equation (1) shows that the flux ( $J_v$ ) is proportional to the difference between the differential operational pressure (which is the difference between the pressures across the membrane) and the differential osmotic pressure (which is the difference between the osmotic pressures across the membrane), i.e.,  $(\Delta P - \Delta \pi)$ .

When the NaCl concentration in an aqueous solution is 1500 ppm, the osmotic pressure is 1.2 kgf/cm<sup>2</sup>, and when the NaCl concentration in a NaCl aqueous solution is 500 ppm, the osmotic pressure is 0.4 kgf/cm<sup>2</sup>. Thus, the flux of a reverse osmosis membrane evaluated with a 500 ppm NaCl solution (which has an osmotic pressure of 0.4 kgf/cm<sup>2</sup>) is about 0.33 (i.e.,  $(5 - 0.4) / (15 - 1.2)$ ) times the value obtained with a 1500 ppm NaCl solution (which has an osmotic pressure of 1.2 kgf/cm<sup>2</sup>). Therefore, a flux of 2.4 m<sup>3</sup>/m<sup>2</sup>·day according to Example 1 of Ikeda et al. is equivalent to 0.79 m<sup>3</sup>/m<sup>2</sup>·day under the test conditions of the present invention. This value is within the

range of the present invention.

On the other hand, with regard to salt rejection, it is necessary to consider both solute flux and volume flux. When the salt rejection ratio is 98%,  $C_2 - C_3 \cong C_2$ . Therefore, the absolute amount of salt passing through the membrane is proportional to the concentration of the solute in the feed solution ( $C_2$ ). The salt rejection ratio can be represented by the following equation:

$$\text{rejection ratio} = (1 - (C_p / C_f)) \times 100 \quad (3)$$

where  $C_f$  is the solute concentration in a feed solution ( $\cong C_2$ ) and  $C_p$  is the solute concentration in a permeate.

The salt (solute) concentration in a permeate ( $C_p$ ) may be obtained by dividing the absolute amount of salt passing through the membrane by the volume of the permeate. Thus, when the operational pressure is reduced while maintaining the salt concentration in the feed solution, the salt concentration in the permeate ( $C_p$ ) becomes higher because the volume of the permeate is reduced. Therefore, the rejection ratio becomes lower when the operational pressure is reduced.

Equation (3) shows that if the salt concentration is changed from 1500 ppm to 500 ppm and the flux remains the same, then there will be no change in the salt rejection ratio. However, when the flux is reduced by a factor of 3, the salt concentration in the permeate ( $C_p$ ) will increase by a factor of 3, which means that  $C_p/C_f$  also increases by a factor of 3. As noted above, the volume flux of the reverse osmosis membrane of Ikeda et al. should be reduced by a factor of 3, if it is evaluated under the test conditions of the

present invention. Therefore, a salt rejection ratio of 98.1% (i.e.,  $C_p/C_f = 1.9\%$ ) of the membrane according to Example 1 of Ikeda et al. is equivalent to a 94.3% ( $C_p/C_f = 5.7\%$ ) salt rejection ratio under the test conditions of the present invention. As noted above, this result is extremely similar to the results shown in the Declaration with reference to Comparative example 1.

Accordingly, membranes of Ikeda do not inherently have the same properties as those of the present invention recited in claim 1. Therefore, Ikeda et al. cannot anticipate or render obvious claim 1. Claims 2 and 5 depend from claim 1 and, therefore, are patentable for at least the same reasons. Accordingly, withdrawal of this rejection is respectfully requested. Similarly, claims 18 and 19 depend from claim 1 and are also patentable for at least the same reasons.

#### **Rejections over U.S. Patent No. 6,171,497**

Claims 1, 2, 5, 18 and 19 stand rejected under 35 U.S.C. § 103(a) as obvious over U.S. Patent No. 6,171,497 ("Hirose").

As a preliminary matter the Applicant notes that by virtue of an assignment recorded at reel/frame No. 010571/0137, the present application and Hirose are commonly owned by the Nitto Denko Corporation. Because the claims of the application (which was filed on December 30, 1999) are only rejected as being obvious over Hirose, and because Hirose only qualifies as prior art under §102(e), the Applicant respectfully asserts that the common ownership exception provided by 35 U.S.C. §103(c) controls. Accordingly, the obviousness rejection of claims 1, 2, 5, 18 and 19 is untenable. Withdrawal of the §103 rejection of these claims is respectfully requested.

In addition, the Declaration provides support for the Applicant's previous assertion that the reverse osmosis membranes disclosed by Hirose do not disclose membranes within the scope of the claims of the present invention.

In Hirose, a reverse osmosis membrane having great surface roughness is treated with an oxidizing agent so that a high flux and a high rejection ratio can be obtained. In Hirose, the membrane has a low flux without the treatment with an oxidizing agent, as evidenced by a comparison between Example 1 and Comparative Example 2. Further, it is shown in Comparative Example 1 that a membrane having minimal surface roughness will only attain a small increase in flux even if it is treated with the oxidizing agent.

Thus, to obtain a reverse osmosis membrane as claimed in Hirose, a membrane with a large surface area (due to great roughness) is treated with an oxidizing agent to further degrade a portion of the surface thereof. Such a reverse osmosis membrane can achieve a high rejection ratio for a charged salt due to repulsion of charged particles. However, it does not have a high rejection ratio for a non-charged solute such as organic substances. This is because the non-charged solute can pass through the membrane from the portion degraded by the oxidizing agent on the surface of the membrane. This is discussed in detail on pages 3 to 4 of the Declaration.

In contrast, to obtain a reverse osmosis membrane as claimed in the present invention, a membrane is formed and then brought into contact with a solution containing an acid halide at a higher concentration. The treatment with the additional acid halide solution increases the acid halide concentration on the surface of the membrane. As a result, more amide and/or carboxylic acid groups are formed on the surface of the membrane, leading to an increased hydrophilicity of the membrane and a high flux.

Unlike the method disclosed in Hirose, the method of forming the membrane according to the present invention does not degrade or cause a defect on the surface of the membrane. Therefore, the reverse osmosis membrane thus-obtained can exhibit a high rejection ratio not only for charged salts, but also for non-charged molecules.

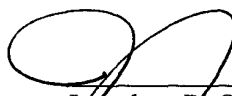


#### IV. Concluding Remarks

Applicant believes this reply to be fully responsive to all outstanding issues and place this application in condition for allowance. If this belief is incorrect, or other issues arise, do not hesitate to contact the undersigned or his associates at the telephone number listed below. Please apply any charges not covered, or any credits, to Deposit Account 50-0591 (Reference Number 04558.039001).

Respectfully submitted,

Date: 7/24/03



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